

# PROBLEMS OF ECOLOGY

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## ENVIRONMENTAL PROBLEMS OF THE FLAME COMBUSTION OF FUEL IN GLASS-MELTING FURNACES

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The environmental characteristics of the flame in glass-melting furnaces are provided. The main harmful emissions released with combustion products and the mechanisms of the formation of some gaseous oxides and dust in the flame are considered. Recommendations for reducing the harmful emissions are supplied.

The main environment-related parameters of glass-melting furnaces are total and harmful emissions released with fuel combustion products, which, on the one hand, are determined by the operating specifics of the furnaces, and on the other hand, by the parameters of flame evolution in the working space of the furnace. The principal harmful emissions released with combustion products are nitrogen oxides, carbon oxide, and dust, and in the case of using fuel oil, sulfur oxides as well. The nature of formation of the above listed oxides is different. It is related to various mechanisms, and in each case the emission concentration in specific conditions is determined by its own regularities. The maximum permissible concentration (MPC) standards for a number of chemicals contained in combustion product emissions from glass-melting furnaces are presented in Table 1 [1].

It should be noted that the environmental regulations for carbon and nitrogen oxides in Russia are considerably stricter than the world standards, the EU and USA regulations, and the norms for sulfur oxides in Russia virtually coincide with the world standards.

The unit value of harmful emissions  $\varepsilon_h$  is closely related to the fuel rate  $b_p$  and the thermal efficiency of the furnace  $\eta_h$  and can be calculated from the following formula:

$$\varepsilon_h = \Psi_h \frac{\Delta q_h}{\eta_h Q_n}$$

where  $\Delta q_h$  is the effective heat rate, kJ/ton;  $Q_n^p$  is the fuel combustion heat, kJ/m<sup>3</sup> or kJ/kg;  $V_\alpha$  is the volume of the products resulting from combustion of a unit of fuel, m<sup>3</sup>/m<sup>3</sup>

or m<sup>3</sup>/ton;  $\Psi_h$  is the concentration of harmful emissions in the combustion products, kg/m<sup>3</sup>;  $v_h$  is the specific value of the total emission, m<sup>3</sup>/ton

Hence obviously

$$b_f = \frac{\Delta q_f}{\eta_h Q_n^p} \quad \text{and} \quad v_h = b_f V_\alpha.$$

Nowadays, in the case of harmful emissions into the atmosphere, the environmental damage is calculated, and the companies are charged payments for environmental pollution based on this calculation. The methods used for evaluation of environmental damage and payments due for that damage involve the value of the total harmful emissions per quarter of year (tons/quarter (year))

$$M_i = \varepsilon_h P_i 10^3,$$

where  $P_i$  is the quarterly or annual production volume, and the index  $i$  corresponds to a particular emission component.

In assessing the environmental damage and the payment due for environmental pollution, the value of reduced emission mass  $M_{ir}$  is used, which is equal to the product of the ac-

TABLE 1

Compound	World standard	EU	USA	Russia
Carbon oxide, mg/m <sup>3</sup>	10–20	–	10	3
Nitrogen oxides, mg/m <sup>3</sup>	0.15	0.135–0.200	0.10	0.04
Sulfur anhydride, mg/m <sup>3</sup>	0.05	0.04–0.06	0.08	0.05

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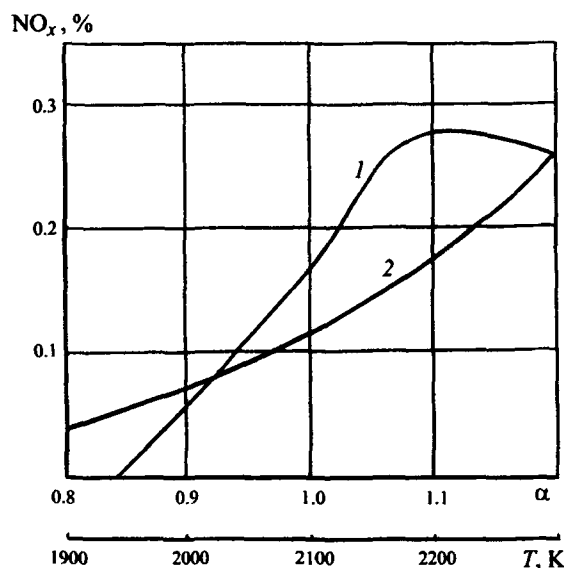


Fig. 1. Dependence of the equilibrium concentration of nitric oxides in combustion products on the air rate coefficient (1) and the temperature in the combustion chamber (2).

tual mass  $M_i$  by the relative aggressiveness factor for the particular emission  $A_i$ :

$$M_{ir} = M_i A_i.$$

The value  $A_i$  is estimated with respect to the average daily aggressiveness of carbon monoxide taken as one. Thus, for example, the relative aggressiveness factor of sulfur anhydride is 22, and that of nitrogen oxides is 41.

Let us consider the specifics of formation of harmful emissions in the furnace flame and analyze ways to reduce them.

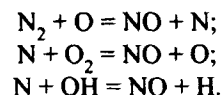
Proceeding from the available experience of operation of different thermal plants and furnaces, the state standard of the Russian Federation for the maximum permissible concentration of  $\text{NO}_x$  in combustion products was adopted (GOST 50591-93). Table 2 gives the maximum permissible concentration values for  $\text{NO}_x$  in the natural gas combustion products at  $\alpha = 1.0$  for regenerative tank glass-melting furnaces heated with natural gas, compared to some other thermal plants.

As can be seen, the maximum standards of  $\text{NO}_x$  concentration for glass-making furnaces are several times greater than the similar standards for other thermal plants. On the

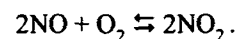
one hand, it suggests that the operating conditions of glass-melting furnaces facilitate the formation of substantial quantities of  $\text{NO}_x$ , and on the other hand, it is indicative of significant reserves for a possible decrease in the level of this type of emissions. It should be noted that the amendments to the GOST 50591-93 standards developed by the JSC Promgaz suggest a decrease in the MPC of  $\text{NO}_x$  to 2000 mg/m<sup>3</sup> for the newly designed glass-melting furnaces.

The studies performed in recent years [2, 3] established that there are three possible ways of NO formation in a hydrocarbon flame: prompt and thermal formation, and formation including intermediate  $\text{N}_2\text{O}$ .

The third way of NO formation is possible only at high pressures. The prompt formation is absent in carbon flame, and its mechanism is not yet sufficiently studied. Therefore, scientists focus their attention on the best studied thermal mechanism of NO formation which was first investigated by Ya. B. Zel'dovich [4]. According to this mechanism, the summary reaction of molecular nitrogen oxidation ( $\text{N}_2 + \text{O}_2 = 2\text{NO} - 186.6 \text{ kJ}$ ) proceeds by the chain mechanism:



The transition of NO to  $\text{NO}_2$  begins at a temperature below 650°C.



The analysis revealed that the conditions facilitating the formation and preservation of nitric oxides include high temperatures, high initial concentration of nitrogen and free oxygen in the gas mixture, an increase in the time of exposing the mixture to a high temperature, and prompt cooling of the mixture. Figure 1 shows equilibrium concentrations of  $\text{NO}_x$  in the combustion products of a methane-air mixture at different combustion temperatures and different air rate coefficients [5]. It can be seen that an increase in the enthalpy of the initial mixture produces an increase in the combustion product temperature and, accordingly, the yield of nitric oxides. Combustion of the fuel mixture with the air rate coefficient significantly greater than 1, as well as a decrease in the combustion temperature brought about by other methods, produce a decrease in the yield of nitric oxides.

TABLE 2

Thermal plants	Rated heat power, MW	Air preheating temperature, °C	Maximum permissible concentration of $\text{NO}_x$ , mg/m <sup>3</sup>
Regenerative tank glass-melting furnaces	7.0 – 40.0	930 – 1100	2500
Continuous heating furnace	7.0 – 65.0	250 – 350	760
Hip roof tube furnaces	3.0 – 80.0	No preheating	600
Hot-water boilers of PTMV type	58.2; 116.3; 209	The same	390

However, the time the gases spend in the actual thermal plants is usually tens of times less than the time required for attaining the equilibrium concentration. Therefore, the empirically obtained constants differ significantly from the rated values. Thus, the constant of the direct reaction of  $\text{NO}_x$  formation in the flame differs 2.7 times from the rated value [3]. Numerous researchers attempt to refine the mechanism of nitric oxide formation proposed by Ya. B. Zeldovich by clarifying and studying the mechanism of the reactions and the meaning of the constants appearing in the calculation formulas, as applied to the specific conditions of flame evolution. Some attempts were made to construct extremely complex chemical kinetics models of  $\text{NO}_x$  formation in methane combustion. In particular, 196 chemical reactions for 32 compounds were accounted, as applied to an isothermic and spatially homogeneous model described in [6]. In all cases, in order to calculate the  $\text{NO}_x$  formation by the thermal mechanism, it is necessary to know the temperature fields in the working space of the furnace, and the variations in the concentrations of combustion product components, especially  $\text{N}_2$  and  $\text{O}_2$ , and also  $\text{NO}$ ,  $\text{H}_2$ , and  $\text{CH}_4$  in the more complex mechanisms. All researchers arrive at the conclusion that the formation of nitric oxides is determined by the temperature level in the local flame volume with an excess concentration of nitrogen and oxygen.

The temperature of air preheating before combustion has a very strong effect on the formation of nitric oxides in the combustion products. A quadratic dependence of  $\text{NO}_x$  concentration on air preheating temperature is typical. Thus, when the air is preheated to  $315^\circ\text{C}$ , the nitric oxide concentration increases 2.5 times compared to the case where the air is not pre-heated. When the air is preheated to  $535^\circ\text{C}$ , the concentration of  $\text{NO}_x$  increases 5 times. It should be noted that extrapolation to preheating temperatures of  $900 - 1000^\circ\text{C}$  gives values of nitric oxide concentration of about  $2500 - 3000 \text{ mg/m}^3$ , which corresponds to the limiting permissible values for regenerative tank furnaces specified in Table 2.

The available data on the change in gas temperature along the length of the flame suggest that shortening of the flame stimulates the formation of  $\text{NO}_x$ . In this case the maximum temperature of the flame increases and shifts to the initial point of the flame space, which increases the time of gas exposure in a high-temperature area. As the flame is shortened, the time of exposure of the gases to the area of relatively low temperatures with relatively low oxygen and nitrogen concentrations decreases. The earlier mentioned factors increasing the yield of  $\text{NO}_x$  (preheating of air, oxygen blowing) simultaneously promote shortening of the flame.

An increase in the flame luminosity increases thermal efficiency, which produces a decrease in the gas temperature and facilitates a decrease in  $\text{NO}_x$  formation.

A powerful factor suppressing nitric oxides is two-phase fuel which produces a decrease in the maximum combustion temperature and leveling of temperature fields in the flame space of the furnace.

An efficient method of decreasing the  $\text{NO}_x$  formation in high-temperature furnaces, including glass-melting furnaces,

consists in the acoustic effect on flame (Russian Federation patent 1455444). The recommended acoustic vibration frequencies are  $1.5 - 2.0 \text{ kHz}$ , and the specific power is  $0.15 - 0.30 \text{ W/m}^3$  of gas volume. The mechanism of this phenomenon is related to the leveling of the flame temperature fluctuations and a decrease in the flame temperature peaks.

Since natural gas virtually does not contain sulfur, its combustion products do not include sulfur compounds. Natural gas that contains  $\text{H}_2\text{S}$ , such as gas from the Orenburg gas field, is subjected to preliminary processing and refinement from sulfur compounds (the Klaus process). However, fuel oil can contain certain quantities of sulfur, and the sulfur content in high-sulfur fuel oil reaches 3.5%. As a result, sulfur dioxide and other compounds appear among the combustion products.

The main method for the reduction in sulfur oxide emission is refinement of the combustion products from sulfur compounds. There are methods involving the introduction of alkaline additives to fuel oil, which suppress the formation of highly toxic sulfur compounds in the combustion products.

The presence of carbon oxide in the combustion products primarily indicates that the fuel combustion process is not properly organized. A prompt chemical analysis of samples of the combustion products at the outlet from the flame space of the furnace, even using the simplest gas analyzer, can ensure required air rate coefficient and length of the flame.

The dust exhaust from the flame space of glass-melting furnaces is determined mainly by technological factors. A radical method for prevention of dust emissions to the atmosphere is gas purification using electric filters. However, the most efficient and rational method of intra-furnace dust precipitation is the use of a turbulizing jet curtain [7] which provides for coagulation of softened dust particles and their intense deposition on the glass melt surface.

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